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THE MANUFACTURE OF VIBRATORILY COMPACTED FUEL ELEMENTS FOR DOPPLER-COEFFICIENT MEASUREMENTS

by

J. E. Ayer, C F. Konicek,
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ABSTRACT

The procedures that are used to manufacture and assemble vibratorily compacted oxide fuels containing plutonium, depleted uranium, and uranium-233 materials are described. Thirty-two acceptable, low-density, fuel rods were made with loadings of 100% PuO_2, 100% $^{233}UO_2$, and various blends of $^{238}UO_2$-PuO_2. The granules for loading were prepared by tableting the respective oxide powder, granulating, firing, and sieving for the proper size range. Over 21 kg of dense, sintered granules were then loaded into the elements by vibratory compaction to densities ranging from 50.8 to 57.8% of theoretical. Fourteen elements were 1/2 in. in diameter and 18 were 1 in. in diameter; the jackets of six elements were made of Invar and the remaining 26 were Inconel.

During in-pile testing, the elements are to be heated by resistance wire that is wrapped around the element. The longitudinal expansion of the elements during heating is accommodated by either a sliding seal or by spring loading on the exterior of the element. Eight of the elements have a sliding seal, whereas the remaining 24 are spring loaded.

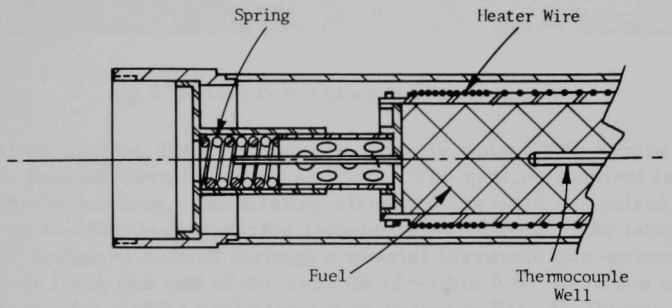
INTRODUCTION

Low-density fuel bodies that are required for Doppler-effect experiments in ZPR-3 were previously produced from pellets of glass-bonded granules of fissionable material.[1] The silicate glass-bonding material maintained voids between the granules and fixed the particles in place. The fuel elements described herein were loaded by vibratory compaction. Vibratory compaction was chosen for simplicity of process and because the vibratorily compacted columns, unlike those formed from pellets, can be regenerated after use. Preliminary investigations[2] showed that the desired

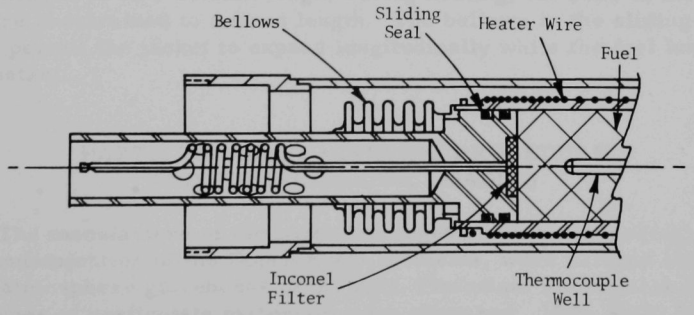
low-density fuel body could be obtained by using a single-component system of particle size range from 50 to 100 mesh. This size range yields an average packing efficiency of about 58% for rounded particles.

DESCRIPTION OF THE FUEL ELEMENT

The Doppler fuel elements consisted of freely expanding elements. Two basic designs were used for accommodating longitudinal expansion, as shown in Fig. 1. One type expanded against an Inconel spring (spring loaded element); in the other design, expansion was accommodated by a sliding seal and a bellows secondary seal (sliding seal element). There were six Invar elements, the remainder being of Inconel. Six specimens had a 0.041-in. wall and contained a 0.400-in.-thick sintered disk, whereas the remainder had a 0.0195-in. wall and a 0.070-in.-thick sintered disk.



Spring Loaded Element



Sliding Seal Element

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Fig. 1. Expansion Accommodation in Doppler Elements

All of the elements were loaded with a 12-in. vibratorily compacted oxide-fuel column. A sliding seal element in loading condition is shown in Fig. 2. The top end cap sits directly on the column of particles and is welded into place.

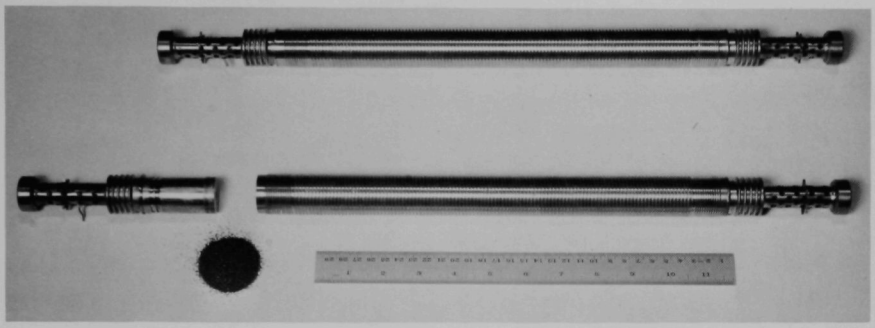

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Fig. 2. Exploded View of Sliding Seal Doppler Element

After loading, the capsules are spirally wound with a mineral-insulated, Inconel-sheath heating element. The heating element is fixed to the capsule by brazing. The heating elements are used to control the temperature of the fuel bodies during Doppler experiments. The temperature of the fuel bodies is sensed through a coaxial thermocouple-protection tube that extends from one end of the capsule to within 0.67 in. of the opposite end. A 1-in.-dia sliding seal element is shown in Fig. 2. During heating, all components of this fuel element tend to expand. To obviate correction due to changes in fuel-column length during heating, the ends of the elements are constrained to a fixed length. The bellows in the sliding seal element permit the jacket to expand longitudinally while the fuel length is held constant.

PARTICLE-MANUFACTURING PROCEDURE AND FUEL-ELEMENT LOADING

The manufacture of the particles, together with the loading, welding, and decontamination of the Doppler fuel elements, were carried out in the helium-atmosphere gloveboxes of the ANL Plutonium Fabrication Facility. Three types of particulate materials were prepared: 100% PuO_2, blends of PuO_2 with depleted UO_2, and 100% $^{233}UO_2$.

The PuO_2 input to the fabrication consisted of eight lots of material prepared by calcination of oxalate by Isochem, Inc., Richland, Wash. Each lot was analyzed by the vendor for plutonium content, flourine, chlorine,

uranium, and carbon. Spectrographic and isotopic analyses were also provided by the vendor. Check analyses were made at ANL, and the results of ANL and vendor analyses are as follows:

Chemical	Assay	$\bar{X}$	n	σ	Analyst
Pu	wt %	86.48	19	0.64	ANL
Pu	wt %	87.58	25	0.29	Isochem
O_2	wt %	12.39	16	0.22	ANL
Cl	ppm	<10	25	-	Isochem
F	ppm	<10	25	-	Isochem
U	ppm	108	25	-	Isochem
C	ppm	517	25	-	Isochem
C	ppm	2500	2	-	ANL
^{239}Pu	wt %	85.944	13	0.201	Isochem
		85.916	3	0.148	ANL
^{240}Pu	wt %	11.470	13	0.183	Isochem
		11.497	3	0.097	ANL
^{241}Pu	wt %	2.405	13	0.047	Isochem
		2.400	3	0.055	ANL
^{242}Pu	wt %	0.181	13	0.006	Isochem
		0.187	3	0.006	ANL

The $^{233}UO_2$ starting material for the fabrication of Doppler elements was obtained from Oak Ridge National Laboratory. Chemical assay and isotopic analyses by ORNL and check analyses by ANL were as follows:

Analysis	$\bar{X}$ (wt %)	n	σ	Analyst
Uranium	86.97	7	1.20	ORNL
Uranium	87.68	2		ANL
^{233}U	98.33	2		ORNL
^{233}U	98.29	1		ANL
^{234}U	1.203	2		ORNL
^{234}U	1.218	1		ANL
^{235}U	0.046	2		ORNL
^{235}U	0.046	1		ANL
^{236}U	0.0038	2		ORNL
^{236}U	0.003	1		ANL
^{238}U	0.423	2		ORNL
^{238}U	0.437	1		ANL

The blends of oxide were made from the above plutonia and depleted uranium oxide supplied by Kerr-McGee Oil Industries, Inc. (Lot 4176-D-1). The chemical analysis on the depleted urania and a typical isotopic analysis for depleted uranium is shown below:

Analysis	wt %	Ratio
Uranium	87.46	
O/U		2.13
^{234}U	0.0014	
^{235}U	0.2089	
^{236}U	0.0058	
^{238}U	99.80	

Beyond the blending procedure for the mixture of materials and the special shielding precautions for the $^{233}UO_2$ materials, the particle manufacturing procedures were identical and as shown in Fig. 3. Briefly, the

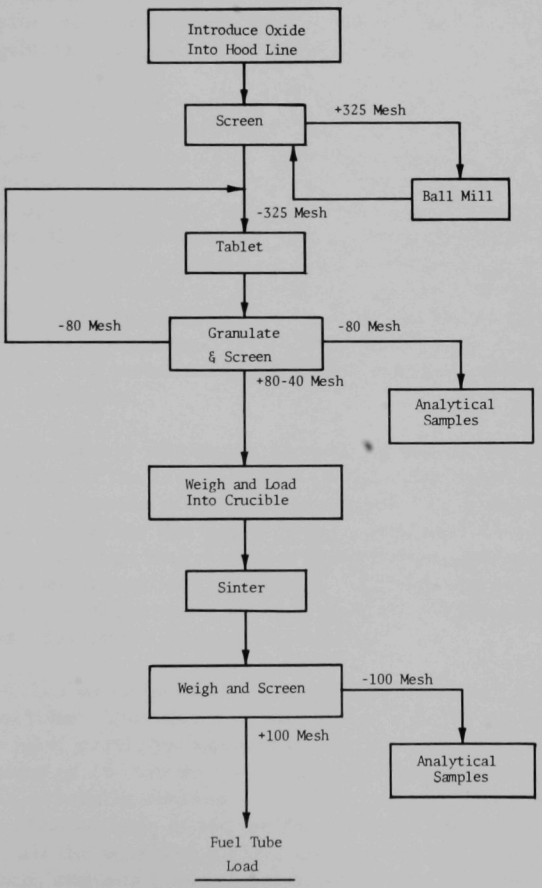

Fig. 3. Flow Diagram of Fabrication of Fuel for Doppler-element Manufacture

blending procedure consisted of (1) weighing proper proportions of PuO_2 and depleted UO_2, based upon metal analyses, and (2) mixing in a vaned tumbler for 24 hr prior to sampling and processing. The special shielding precautions for the production of $^{233}UO_2$ granules are described in a separate section of this report. No milling of the materials was required because of their fineness.

Batches of the powdered materials weighing about 20 g were pressed into 1.0-in.-dia tablets at a pressure of 15,000 psi. This produced tablets about 0.8 cm thick at about 44% of theoretical density. The tablets were then granulated and classified by pushing them through a 40-mesh screen (420-μ opening) and collecting the particles on an 80-mesh screen (177-μ opening). Samples were then taken for oxygen and metal analyses in anticipation of determining changes caused by sintering.

Batches of granulated material weighing 1 kg were loaded into yttria-stablized zirconia crucibles and fired for about 1 hr at 1650°C under a helium atmosphere of 20-25 in. Hg absolute pressure. The lightly caked sintered material was removed from the crucible and pressed through a 50-mesh (297-μ opening) screen and collected on a 100-mesh (149-μ opening) screen. The reduction of the screen opening sizes (between granulation of unfired and fired particles) compensated for shrinkage during sintering. About 6% of the sintered material passed through the 100-mesh screen and was not used. The density of the sintered material was determined by pycnometry using bromobenzene. The sintered and screened material was sampled for oxygen and metal analyses to determine furnace losses and to characterize the particles.

Fuel-rod assembly was begun by welding the bottom cap in the fuel tube. During loading of the fuel material, it was important to prevent alpha contamination of the outside of the fuel specimen. To prevent contamination, the fuel element, except for the upper 3/4 in., was enclosed in shrinkable plastic tubing. Teflon tape was wrapped around the upper exposed end and this end was clamped into a loading fixture. An element ready for loading is shown in Fig. 4. A Nichrome wire was used to cut the plastic tubing for its removal upon completion of particle loading.

The particles were loaded through a plastic funnel to the desired height in the fuel tube. The vibrator was run at a force of 2.5 g at 60 Hz for periods of 3 min; particles were added until settling ceased. Usually, a total vibration time of 15 min was sufficient to complete the loading. The capsule was then carefully cleaned on the top 1/4 in. of the inside diameter of the fuel tube. The outside of the Teflon tape and plastic tubing was also cleaned; finally, all the tape and plastic tubing was removed. The end cap was taped in place, and any loose alpha activity was cleaned from the outside of the fuel tube.

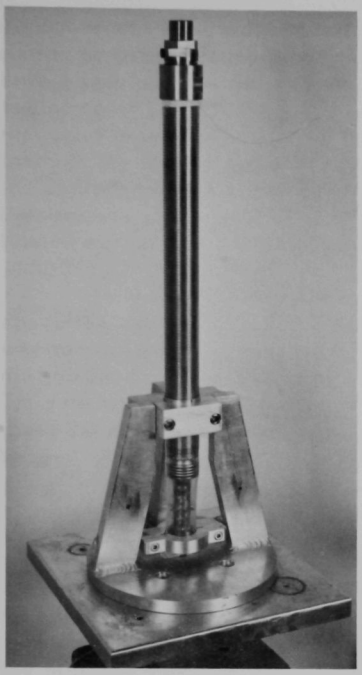

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Fig. 4. Doppler Element in Loading Fixture

The top end cap was welded in place, and the elements were tested for leak tightness. All of the elements were leak tested by evacuation to 5 psi through the capillary tube and monitoring for a pressure rise. If no pressure rise occurred, the elements were considered satisfactory and the capillary tube was pinched shut; the excess tubing was trimmed off, and the trimmed end was fused shut. Finally, the elements were leak checked by helium mass spectrometry by inserting the entire element into a vacuum chamber at room temperature. All elements were acceptable and showed a leak rate, at the limit of detection, of less than 3.7×10^{-8} std cc/sec of helium with an exterior pressure of less than 10^{-5} mm Hg.

The fuel rods were then checked for alpha contamination and cleaned until the loose activity was less than 10 disintegrations/min and fixed activity less than 600 disintegrations/min. Cleaning was generally done with dry wipes, but occasionally light abrasion of a weld with an electrically operated rubber eraser was required. Upon completion of cleaning, the fuel rods were accumulated in storage pending the heater wire-wrapping operation.

FABRICATION OF $^{233}UO_2$

The major problem in processing ^{233}U fuels nonremotely is the penetrating radiation hazard to operating personnel. Residual ^{232}U impurity, whose daughter products produce significant beta and gamma radiation, is responsible for the radiation hazard.

Six $^{233}UO_2$ fuel elements were fabricated by loading of 3.5 kg of granules. The fabrication of the elements was done in two main phases: granule preparation, and loading, welding, and testing. Granule preparation was the most time-consuming and thus provided the greatest radiation hazard. Since this operation took place mainly in one glovebox, it was convenient to use local shielding to reduce the radiation level to tolerable levels. An 1/8-in.-thick lead sheet with a 1/4-in. leaded glass

viewport was hung on the face of this glovebox. Further shielding was obtained by using leaded gloves. Even though this batch of $^{233}UO_2$ contained only 4 ppm of ^{232}U, the radiation levels were still substantial. The primary radiation of 1.3 R through the container vessel was reduced to 90 mrem through the lead sheet, to 55 mrem through the viewport, and to 80 mrem through the leaded gloves. Local shielding, around the material, reduced the radiation level further to 20 mrem through the lead, and 18 mrem through the glass. An additional precautionary measure was the use of a leaded apron.

To monitor the personnel radiation dosage, film badges were worn above the eyes and on the fingers and wrists. The normal whole-body dose was monitored by a badge worn on the shirt pocket. The permissible radiation levels were 100 mrem to the eyes, 200 mrem to the body, and 1.0 rem to the extremities per 40-hr work week. In a few isolated cases, these levels were modestly exceeded for the eye badge and the whole-body badge.

RESULTS

Twenty-one different firing runs (see Table I) were made as follows: nine with plutonia, nine with mixed oxide, and three with $^{233}UO_2$. There was a sufficient number of runs so that a statistical analysis could be made for the plutonia data, the $^{233}UO_2$ data, and the mixed-oxide data. The average values ($\bar{X}$) and standard deviations (σ) for these materials are summarized for each compound and mixture in Table I. The variation in the data for the nonblended compounds represents sampling and analytical error, whereas the blended material additionally includes error due to nonhomogeneity or imperfect blending.

The analyses of PuO_2 provided by Isochem, Inc. showed a standard deviation of 0.29 in the plutonium content between batches. The ANL analyses of firing batches show, in Table I, a standard deviation of 0.32 in plutonium content. The agreement between these two figures indicates that all of the 100% PuO_2 elements may be treated as a single composition, with 95% confidence, of 88.25 ± 0.64% plutonium. A similar analysis of ^{233}U in fired granules showed a standard deviation of 0.27 in uranium content between batches. This establishes 95% confidence in uranium content of 87.65 ± 0.54 for all elements containing ^{233}U.

The variation in analyses for plutonium in PuO_2 and uranium in UO_2 indicates that analytical error and batch-to-batch variations introduce a standard deviation of about 0.3% of the metal content. If the assumption is made that this error is consistent, one may assign a deviation due to inhomogeneity and blending errors in the blended elements. The total variance in analyses for a blend (σ_t^2) is assumed to consist of analytical error

(σ_a^2) and variations due to inhomogeneity (σ_h^2); i.e., $\sigma_t^2 = \sigma_a^2 + \sigma_h^2$. Then, the standard deviations of metal composition due to inhomogeneity (σ_h) are determined as $\sigma_h = \sqrt{\sigma_t^2 - \sigma_a^2}$. Table II shows the total standard deviation, the standard due to analytical error, and the calculated standard deviation due to inhomogeneity for all firing batches encapsulated in the Doppler elements described.

TABLE I. Results of Analytical Chemistry and Statistical Analysis of Fired Granules for Doppler Capsules

Firing Batch No.	Analytical Results			Elemental Ratios		Special Materials Batch Identity
	Plutonium	Uranium	Oxygen	U/Pu	O/M	
1	-		11.36	0	1.92	SO26-5-1
			11.39			
2	-		10.92			SO26-5-6
			10.97			
3	88.74		11.40			SO26-5-7
	88.60		11.36			
	88.23					
4	87.97		11.32			SO26-5-38
	87.81		11.22			
5	-		11.50			SO26-5-43
			11.48			
6	-		11.48			SO26-5-44
			11.40			
7	88.18		11.45			SO26-5-46
	88.24		11.46			
8	-		-			SO26-5-47
9	-		11.30			SO26-5-48
			11.35			
n	7		16			
$\bar{X}$	88.25		11.33			
σ	0.32		0.17			
10	40.36	45.90	10.29	1.00	1.97	1-61-8-1711/A
	40.61	45.86	12.84			SO26-5-4
11	-	-	11.47			2-61-8-1711/A
			11.51			SO26-5-4
12	45.46	42.47	11.75			1-61-8-1711/B
	45.37	42.23	11.37			SO26-5-17
	45.09	42.36				
	44.87	42.29				
13	43.18	43.74	11.34			2-61-8-1711/B
	43.00	43.68	11.46			SO26-5-17
	43.23	43.67				
	43.44	43.87				
n	10	10	8			
$\bar{X}$	43.46	43.61	11.50			
σ	1.84	1.37	0.17			
14	10.98	76.33	11.47	7.10	2.00	1A-61-8-1701
	10.86	76.30	11.70			SO26-5-40
	10.98	76.36				
	10.92	76.20				
15	10.78	76.40	11.72	7.10	2.00	1B-61-8-1701
	10.80	76.20	11.71			SO26-5-40
	10.77	76.41				
	10.75	76.30				
16	10.49	75.61	11.73			1C-61-8-1701
	10.63	75.88				SO26-5-40
	10.59	75.78				
	10.66	75.55				
n	12	12	5			
$\bar{X}$	10.77	76.11	11.67			
σ	0.15	0.32	0.17			
17	28.79	57.77	11.53	2.01	1.98	61-8-1702
	28.78	57.58	11.48			SO26-5-40
	28.95	58.01				
	28.81	57.76				
n	4	4	2			
$\bar{X}$	28.83	57.78	11.51			
σ	0.08	0.18	-			
18	58.64	29.26	11.22	0.50	1.91	61-8-1703
	58.47	29.10	11.18			SO26-5-40
	58.52	29.12				
	58.28	29.21				
n	4	4	2			
$\bar{X}$	58.48	29.17	11.20			
σ	0.15	0.08	-			
19		87.90	-	∞	2.06	SSNDFC-2-1
		87.86				SSNDFC-2-3
		87.76				
		87.87				
		87.90				
20		87.57	12.20			SSNDFC-2-6
		87.42	12.31			SSNDFC-2-7
		87.90	12.50			
		87.37	12.29			
21		87.60	12.20			SSNDFC-2-8
		87.02	12.57			SSNDFC-2-9
		87.51	12.60			
		87.76				
n		13	7			
$\bar{X}$		87.65	12.38			
σ		0.27	0.17			

TABLE II. Error Analysis of Fired Granule Compositions

	Plutonium				Uranium				Oxygen		Average Ratios	
	$\bar{X}$	σ_t	σ_a	σ_h	$\bar{X}$	σ_t	σ_a	σ_h	$\bar{X}$	σ_a	U:Pu	O:M
100% PuO_2	88.25	0.32	0.32	-	0	-	-	-	11.33	0.17	0	1.92
1 UO_2:2 PuO_2	58.48	0.15	0.21	0	29.17	0.08	0.09	0	11.20		0.50	1.91
1 UO_2:1 PuO_2	43.46	1.84	0.16	1.83	43.61	1.37	0.13	1.36	11.50		1.00	1.97
2 UO_2:1 PuO_2	28.82	0.08	0.11	0	57.78	0.18	0.18	0	11.51		2.01	1.98
7 UO_2:1 PuO_2	10.77	0.15	0.04	0.14	76.11	0.32	0.23	0.22	11.67		7.10	2.00
100% UO_2	0	-	-	-	87.65	0.27	0.27	-	12.38		∞	2.06

Table II permits one to establish confidence limits for the metal contained in all Doppler capsules of a particular composition. In the event that each capsule must be treated individually, Table III presents complete loading data for each Doppler capsule, and lists the amount of material and the source firing batch number for each capsule. Particle densities were obtained by the pycnometer method using bromobenezene as the fluid. The theoretical density is calculated on the basis of the isotopic content of the material and the type of material, whether it is urania, plutonia, or a blend. The fuel density is the weight of material divided by the volume of each fuel element, the packing efficiency is the parameter that shows the percentage of the total fuel cavity occupied by the particles. The packing efficiency depends mainly on the particle size and shape and is not dependent on particle density, so that it is the most reproducible fuel loading number from capsule-to-capsule. The packing efficiency varied from 54.5 to 62.6%, as expected, for one size fraction varying in size from 50 to 100 mesh.

TABLE III. Loading Data for Oxide Particles in Doppler Capsules

Capsule Number	Contents		Jacket			Loading				
	PuO_2, %	UO_2, %	ID, in.	Material	Type	Weight, g	Packing Efficiency, %	Fuel Density, g/cc	Theoretical Density, %	Firing Batch No., wt
INV-1[a]	100	0	1	Invar	Sliding Seal	658	-	-	-	-
2[a]			1			1067	-	-	-	-
3[a]			1			1059	-	-	-	-
4[a]			1			965	58.1	6.15	53.7	3, 953 g; 7, 12 g
INC-1			1	Inconel		1066	57.9	6.19	51.2	7, 947 g; 8, 119 g
2			1			1066	58.8	6.23	54.3	8, 772 g; 6, 294 g
3			1		Spring Loaded	1012	56.6	6.03	52.6	9, 178 g; 1, 834 g
4			1			1023	57.3	6.10	53.3	1, 49 g; 5, 954 g; 4, 20 g
5			1			1041	57.7	6.20	54.1	4, 916 g; 2, 125 g
6			1/2			239	62.3	6.51	56.8	6, 239 g
7			1/2			243	62.6	6.55	57.1	6, 243 g
8			1/2			239	61.6	6.44	56.2	6, 199 g; 9, 40 g
9			1/2			239	61.8	6.46	56.5	9, 239 g
10			1/2			240	60.8	6.43	56.0	9, 240 g
11			1/2			240	61.5	6.52	56.8	9, 240 g
12	67	33	1			1006	56.6	5.93	52.6	18, 1006 g
13	50	50	1			976	54.5	5.80	51.7	11, 956 g; 10, 20 g
14	50	50	1			1003	56.0	5.95	53.1	12, 865 g; 10, 138 g
15	50	50	1/2			221	55.4	5.91	52.8	10, 221 g
16	50	50	1/2			223	55.7	5.95	53.1	10, 223 g
17	50	50	1/2			228	56.9	6.10	54.3	10, 228 g
INV-5[a]	50	50	1	Invar	Sliding Seal	970	58.6	6.23	55.6	10, 26 g; 13, 944 g
INC-26	33	67	1	Inconel	Spring Loaded	1065	59.1	6.32	56.8	17, 1065 g
INC-18	12.5	87.5	1			1028	57.5	6.10	55.3	14, 450 g; 15, 578 g
19	12.5	87.5	1			1079	59.9	6.37	57.8	14, 79 g; 15, 40 g; 16, 960 g
25	12.5	87.5	1/2			221	55.8	5.91	53.7	14, 221 g
INV-6[a]	0	100 $^{233}UO_2$	1	Invar	Sliding Seal	914	54.7	5.78	53.8	19, 914 g
INC-20	0	100 $^{233}UO_2$	1	Inconel	Spring Loaded	937	56.3	5.60	52.1	19, 185 g; 20, 752 g
21			1/2			203	55.0	5.46	50.8	20, 203 g
22						204	55.0	5.50	51.2	20, 189 g; 21, 15 g
23						215	57.2	5.78	53.8	21, 215 g
24						212	55.5	5.68	52.8	21, 212 g

[a]0.041-in. wall and 0.400-in.-thick sintered disk; all the other elements had a 0.0195-in. wall and a 0.070-in.-thick sintered disk.

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